

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Appl No. : 10/539,789 Confirmation No. 5011

Applicant : Pfeiffer et al.

Filed : April 3, 2006

Title : Direct synthesis method for the production of etherified melamine resin condensates, melamine resin condensates and use thereof

TC/A.U. : 1796

Examiner : Listvoyb, Gregory

Attorney Docket No. : 4385-051182

DECLARATION UNDER 37 CFR § 1.132

I, **Markus Machherndl**, hereby declare that:

1. I am an expert of the subject matter disclosed and claimed in the subject patent application, U.S. Patent Application Serial No. 10/539,789.

I received a Master's degree in Chemistry from TU Graz (Austria) in 1998 in the field of chemical engineering. Since then I have been working in Research and Development departments. I have been working for 6 years in the field of flame retardancy of polypropylene with melamine based flame retardants (2 Patents), and development of new products based on melamine resin chemistry like resins for WPC application, injection moulding application, SMC-application and fibre application. This was followed by 3 years of research in the field of scale up of lab-processes into pilot plant size.

For the last 1.5 years, I'm working in the area of down scaling C₂ to C₄ hydrocarbons (ethylene, propylene, butene). My actual working field includes pilot plant operations, process development of metathesis of ethylene and 2-butene, dimerisation of ethylene, and dehydrogenation, as well as further development of the existing fibre process of melamine resins. **Currently, I am employed by Borealis Polyolefin GmbH. Borealis Agrolinz Melamine GmbH is a sister company in the same group called Borealis AG.**

I am a designated inventor of a number of patents and/or patent applications including US 2006/0100317 A1, US 7,196,131 B2, US 7,012,108 B2, AT 411.902, EP 1 603 969 B2, EP 2 044 135 A1, WO 2009/027385 A1 and US 2008/0227889 A1.

2. I have reviewed the above-identified patent application, the pending claims thereof, the Office Action of August 18, 2009 and cited references, including the rejections of: claims 25-34 and 37-48 under 35 U.S.C. §103(a) as being obvious over U.S. Patent Publication No. 2004/0024131 A1 ("Borner et al.") in view of U.S. Patent No. 2,473,463 ("Adams") and U.S. Patent No. 4,271,286 ("Michel et al."), and claim 38 as being obvious over Borner et al. in view of Adams and Michel et al., further in view of U.S. Patent No. 5,206,066 ("Horacek") as set forth in the Office Action for the above-identified patent application. I have reviewed and considered the cited prior art as set forth in the Office Action, and I believe that none of the cited prior art, combined as set forth in the Office Action, discloses or suggests a process such as that of present claim 25 comprising, *inter alia*, further concentrating a pre-condensate up to 95-99 wt% in a second vaporization step, and adding a C₄-C₈ alcohol, diols or tetrahydric alcohols to the concentrate after the second vaporization step.

3. I am familiar with the disclosure of Borner et al. Borner et al. discloses a synthesis process comprising the following steps:

- synthesis of an unetherified melamine resin,
- etherification of the melamine resin with methanol under acidic conditions,
- stabilisation of the formed resin by adding a base in MetOH or desaltification by adding butanol,
- procuring the resin in an extruder.

A specific process is described in Example 1 of Borner et al., having the steps of:

- a) forming an etherified melamine resin pre-condensate (melamine, formaldehyde, methanol) and neutralising the pre-condensate solution with KOH in methanol,
- b) concentrating the pre-condensate by distilling off 7 l methanol,
- c) adding 2 l butanol for salt precipitation, and
- d) re-concentrating the resin, and
- e) reaction in an extruder.

The order of the above steps can thus be summarized as etherification - concentration - alcohol addition - concentration - extruder reaction.

The resins of the Borner et al. process can be processed as a melt, have softening points of above 100 °C and are curable above 240 °C. Furthermore, the obtained resins are not free of hydroxymethylenc-amino groups and NH-CH₂-O-groups, as already has been acknowledged by the Examiner.

4. In contrast to Borner et al., the present claim 25 is directed to a process for preparing etherified melamine resins with weight-average molecular weights from 500 to 50,000 free from hydroxymethylenc-amino groups bonded to the triazine rings of the melamine resin condensate and from -NH-CH₂-O-CH₂-NH-groups linking triazine rings comprising the following steps:

- a) forming an etherified melamine resin pre-condensate (melamine, formaldehyde, alcohol),
- b) concentrating the pre-condensate up to 65-85 wt% in a first vaporization step,
- c) further concentrating the pre-condensate up to 95-99 wt% in a second vaporization step,
- d) adding a C4-C8 alcohol, diols or tetrahydric alcohols to the concentrate after the second vaporization step, and
- e) reacting the mixture in an extruder.

The order of the steps according to present claim 25 can thus be summarized as follows: etherification - 1st concentration - 2nd concentration - alcohol addition for transesterification - extruder reaction.

Thus, the present claim 25 differs in the order of the process steps from the teaching of Borner et al. The most striking difference is the addition of alcohols to a precondensate with a concentration of 95 to 99 wt% after the concentration has been completed.

Assuming arguendo that a person skilled in the art would even recognize the re-concentration step in example 1 of Borner et al. as the second vaporization step, then the

addition of butanol as alcohol in step c) according to Borner et al. is definitely carried out before this second vaporization step.

It is also to be pointed out, that the pH mentioned in paragraph [0030] of Borner et al. does not refer to the pH as applied in an etherification process, i.e., methylolation and etherification of the melamine-formaldehyde resin, but rather to the stabilisation process of the etherified resin. This is a different step in the overall synthesis process, in contrast to the statement by the Examiner on page 3, 5th paragraph of the Office Action.

Furthermore, the conclusion by the Examiner on page 4, 2nd paragraph of the Office Action that the pelletized resin suggests a Molecular weight of at least 500 lacks a technical basis. It is not possible to assume a certain molecular weight of a resin from the fact that a resin can be pelletized. For example, pelletized resins with very low molecular weights (< 200 g/mol corresponding to 1 mol Melamine and 1-2 mol formaldehyde) are known. On the other hand, high molecular weight resins with molecular weights of > 2000 g/mol can be liquid or pasty which can by no means be present in any pelletized form.

Furthermore, the Examiner states on page 4 of the Office Action that Borner et al. does not disclose any solid content after vaporization. However, the Examiner argues that the temperature conditions in the extruder between 160 and 185°C according to Borner et al. are sufficient to obtain a dry resin having a solid content of 95 to 99 wt%. In my opinion, I kindly disagree with the Examiner's contention that the temperature conditions in the extruder between 160 and 185°C according to Borner et al. are sufficient to obtain a dry resin having a solid content of 95 to 99 wt%.

According to the present process, the highly concentrated melt is still meltable and in a non-cured state after the 2nd vaporization step. Only the temperature conditions in the extruder in the final process provide a cured resin. Thus, the temperature conditions in the extruder according to Borner et al. cannot serve for arguing that a highly concentrated resin in the first reaction steps is present before alcohol addition and before extrusion. On the contrary, if only after the treatment in the extruder a dry and highly concentrated resin is obtained (according to the Examiner's arguments), this would then mean that the resin must have been less concentrated before the treatment in the extruder. In contrast, according to the present process, a highly concentrated resin is already obtained before the extrusion process

and not during the extrusion process. Hence, the resin according to Borner et al. must imperatively be different to the resin obtained by the present process.

5. Adams describes a synthesis process comprising the following steps:

- synthesis of a melamine formaldehyde precondensate with low formaldehyde content
- etherification of the melamine resin with methanol and simultaneous alcohol distillation,
- concentration of resin by heating,
- trans-etherification with butanol and/or propanol and simultaneous alcohol distillation
- concentration of resin by heating up to 85% of solids content.

Example 3 (column 6, starting line 56 of Adams) describes for instance the following process:

- mixing of melamine and formaldehyde with not more than 3,5 mole formaldehyde per mole of melamine,
- continuous addition of methanol under simultaneous distillation of alcohol and water providing an etherified resin,
- continuous addition of n-butanol under simultaneous distillation of alcohol and water providing an transetherified resin,
- heating the mixture under reduced pressure for concentration of the liquid resin.

Thus, only after the final product in the form of a liquid composition is obtained said product might be concentrated to yield products comprising 10-85 wt% of resin (column 5, lines 1-5, Examples 3 and 5). Adams does also not disclose that an alcohol is added after two resin concentration steps, in contrast to the presently claimed invention.

The order of reaction steps according to Adams is therefore: etherification - alcohol addition - transetherification - concentration.

The process of Adams provides a liquid etherified melamine resin with low formaldehyde content, which is usable as lacquer, varnish, for films, coatings or impregnation (column 9, lines 15-22 of Adams).

In my opinion, the general teaching of Adams is that an etherification or trans-etherification reaction of a melamine formaldehyde resin with low formaldehyde content only

works successfully in good yields by distilling the excess alcohol and water simultaneously to the alcohol addition. As the distillation proceeds, the alcohol has to be added from time to time until completion. Hence the "two step"- distillation (see Examiner's comment on page 6 of the Office Action) in the Adams process does not serve to increase the concentration of the melamine resin but rather serves to promote the etherification reaction and subsequently the trans-etherification reaction.

In my opinion, a person skilled in the art would not consider Adams when looking for a process as to how to produce melt processable melamine formaldehyde resins, since Adams deals solely with liquid aqueous resin solutions usable in lacquer and coating industry.

If, nevertheless, a person skilled in the art would consider the teaching of Adams for modifying the resins, he/she still would not arrive at the present synthesis process. Although it is correct that Adams discloses fully etherified (and trans-etherified) melamine formaldehyde resins with low formaldehyde content free of hydroxymethyleneamino groups and NH-CH₂-O-groups, Adams – as in Borner et al. - does not disclose a two step resin concentration up to 95-99 wt% resin content with subsequent addition of the alcohol such as is presently claimed.

6. Michel et al. describes a two step synthesis process for production of alkanol etherified melamine formaldehyde resins in which the extent of the etherification is 30 to 60 %, having the following steps:

- reaction of melamine with formaldehyde and water and 0-30 wt% of an alkanol at 60-90 °C and pH = 8-11, then
- addition of the remaining alkanol at 80-130 °C and pH = 3-7 in presence of strong acids providing an etherified resin, then
- concentration of the resin up to a solids concentration of 75 wt%.

The order of reaction steps according to Michel et al. is therefore:
methylation- alcohol addition -etherification - concentration.

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The process of Michel et al. provides liquid etherified melamine resins with good compatibility with organic binders, low viscosity, good reactivity and storage stability, which is usable as lacquer, varnish for coatings, adhesives or laminates.

In my opinion, Michel et al. is solely directed towards liquid aqueous resin solutions usable in the lacquer and coating industry. Therefore, a person skilled in the art would not consider Michel et al. when looking for a method for producing melt processable melamine formaldehyde resins.

Michel et al. (equally to Borner et al. and Adams) does not disclose a two step vaporization process for obtaining a resin with a concentration of 95 to 99 wt% and a subsequent alcohol addition, such as is presently claimed. As shown in the examples of Michel et al., the concentration increase of the final resin is carried out only after completion of the etherification reaction, hence after the whole amount of the alkanol has been added to the mixture and after the reaction mixture already has been cooled down from reaction temperature. Furthermore, the maximum resin concentration disclosed by Michel et al. is only 75 wt%. When considering the teaching of Michel et al., one skilled in the art would come to the conclusion that it is not even desirable to produce a resin with a still higher solids concentration. As shown in column 2, line 35 of Michel et al., only products with a preferably low viscosity are desired. Since it is common knowledge that by increasing the solids content the viscosity of the resin also increases, the teaching of Michel et al. even leads the expert away from the invention.

The Examiner's argument at page 7 of the Office Action is not pertinent to the desired use of the resin. In the special technical field of the invention - namely production of melt processable melamine resins- the advantages of a high solids content are beyond doubt (see also below).

7. The Examiner is of the opinion that it would have been obvious to a person skilled in the art to obtain a 95-99 wt% melt of etherified melamine resin precondensate over Borner et al. in view of Adams and Michel et al. I kindly disagree.

In my opinion, increasing the concentration of a precondensate melt over 85 wt%, especially up to 95-99 wt%, is **not** a trivial and obvious process. In order to obtain a

melt having a concentration over 85 wt%, various technical aspects have to be considered. The concentration process is in general accompanied by an extreme foam formation and viscosity increase. Therefore, the first vaporization step is carried out in a thin-film vaporizer with a rotational speed of 1400 U/min which is applicable for melts having low viscosities.

In the second vaporization step, however, only a vaporizer with a lower rotational speed of 400 U/min can be applied. Such a slow vaporizer acts as a foam breaker and thus allows a further concentration of the melt up to 95-99 wt%. At the exit of the second vaporizer the melt is almost free of solvent. This is also absolutely required for the further work up of the melt in the extruder. Otherwise the melt would not reach the reaction temperature above 200°C in the extruder in a timely manner required for the build up of the molecular weight.

This means, in reverse, if the second vaporization step is omitted and the melt enters the extruder only with a concentration of less than 85 wt% (or 75 wt% according to Michel et al.) repeated solvent evaporation occurs in the extruder. This in turn would cause a delay of the temperature increase to the required reaction temperature in the extruder. This also would require more energy and extruders with an enormous length in order to obtain the required residence time.

Thus, it is **not** obvious for a person skilled in the art to obtain melt with concentrations higher than 85 wt%. This can also be deduced from the fact that none of the cited references discloses a melt having a concentration above 85 wt%.

The advantages of adding an alcohol for transesterification after the concentration-increase process are as follows: After the second concentration step the melt exists as a syrupy melt with a concentration of up to 99 wt%. The paste is metered via a feeding pump and a feeding line into the extruder (or kneader) where the molecular mass built up and procuring occurs. By adding the alcohol to the melt at this stage the viscosity of the syrupy melt is decreased. This in turn reduces the adhesion of the melt to the wall of the feeding line and therefore the transport of the melt in the feeding line is improved.

Furthermore, the addition of the alcohol to the highly concentrated melt also lubricates the feeding pump so that the addition of an otherwise necessary lubricant is

avoided, e.g., the addition of graphite as lubricant would decrease the procuring and product quality. If the alcohol would be added **before** the second vaporization step the addition of a lubricant would be required.

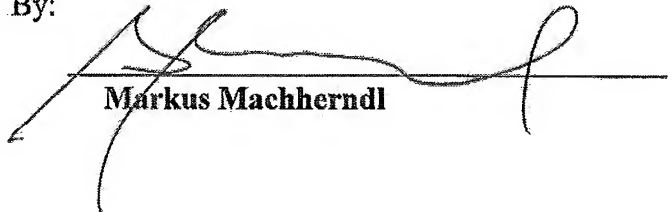
A good mixing of the syrupy melt and the alcohol is achieved by metering the alcohol into slot of the sliding ring of the gear wheel feed pump. If the alcohol is added at a different process step, e.g., before the second vaporization step, a static mixer would be required in order to obtain a good mixing result. Static mixers however are characterized by certain dead zones which can promote blocking in the feed line between the feeding pump and the extruder.

Furthermore, the addition of colder alcohol to the melt having a temperature above 120°C effects a cooling of the melt. Cooling is desirable since it promotes foam breaking. However, the cooling effect is so marginal that a fast heating of the melt in the extruder is not hampered.

8. I declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Executed this on 2nd Decem^{ber} 2009

By:


Markus Machherndl